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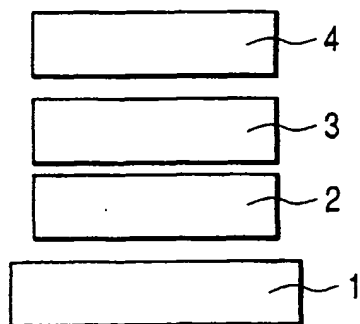
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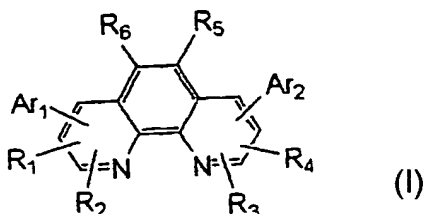
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(54) Title: PHENANTHROLINE COMPOUND AND ORGANIC LIGHT EMITTING DEVICE USING SAME



(57) Abstract: A novel phenanthroline compound is provided which is represented by the general formula [I]: (wherein R<sup>1</sup>#191, R<sup>2</sup>#191, R<sup>3</sup>#191, R<sup>4</sup>#191, R<sup>5</sup>#191 and R<sup>6</sup>#191 are the same or different and each is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, and a halogen atom, and Ar<sup>1</sup>#191 and Ar<sup>2</sup>#191 are the same or different and each is selected from an unsubstituted or substituted fluorenyl group, an unsubstituted or substituted fluoranthenyl group, an unsubstituted or substituted perylenyl group, and an unsubstituted or substituted carbazolyl group). An organic light emitting device using the phenanthroline compound is also provided that has a light output with a high efficiency and a high luminance and has a high long-term durability.



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*For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.*

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## DESCRIPTION

PHENANTHROLINE COMPOUND AND ORGANIC LIGHT EMITTING  
DEVICE USING SAME

5

## TECHNICAL FIELD

The present invention relates to a novel organic compound and an organic light emitting device using the same.

10

## BACKGROUND ART

An organic light emitting device is a device in which a thin film containing a fluorescent organic compound or a phosphorescent organic compound is interposed between an anode and a cathode; excitons of the fluorescent compound or the phosphorescent compound are generated by injection of electrons and holes from the electrodes and a light radiated when the excitons return to the ground state is utilized.

20

In a research by Eastman Kodak Company in 1987 (non-patent document 1), there is reported a light emission of about 1,000 cd/m<sup>2</sup> at an applied voltage of about 10 V for a device of functionally separated two-layer structure using ITO for an anode and a magnesium/silver alloy for a cathode, respectively, an aluminium-quinolinol complex as an electron-transporting material and a light emitting material

25

- 2 -

and a triphenylamine derivative as a hole transporting material. Related patents include patent documents 1 to 3.

In addition, light emission of from ultraviolet region to infrared region is possible by changing the type of the fluorescent organic compound and researches of various compounds have been conducted actively recently. For example, they are described in patent documents 4 to 11.

10 In recent years, there have been a number of studies in which phosphorescent compounds are used as a light emitting material and an energy in a triplet state is used for an EL (electro luminescent) emission. A group of Princeton University has  
15 reported that an organic light emitting device using an iridium complex as a light emitting material exhibits a high light emission efficiency (non-patent document 2).

Moreover, a group of Cambridge University has  
20 reported (non-patent document 3) an organic light emitting device using a conjugated polymer other than the organic light emitting device using the low-molecular materials as described above. In this report light emission in a monolayer is confirmed by  
25 forming a film of polyphenylenevinylene (PPV) in a coating system.

Related patents on organic light emitting

devices using conjugated polymers include patent documents 12 to 16.

Thus, recent progress in organic light emitting devices is remarkable, and possibilities for a wide  
5 range of applications are indicated since it is characterized in that a thin and lightweight light emitting device having a high luminance at a low applied-voltage, diversity of light emitting wavelength and high-speed response can be prepared.

10 However, a higher-luminance light output or high conversion efficiency is required under present circumstances. In addition, there are numbers of problems in terms of durability such as variation with the elapse of time during use for a long period  
15 of time and the deterioration due to an atmospheric gas including oxygen or humidity. Moreover, the light emission of blue, green and red having a good color purity is required for applications such as a full-color display, but these issues are not  
20 sufficiently satisfied.

On the other hand, phenanthroline compounds are used as an electron transporting material or a light emitting material by the excellent electron  
transporting property thereof. Examples of documents  
25 in which the phenanthroline compounds are reported to be used for an organic light emitting device include patent references 17 to 21, but their properties when

they are used as a light emitting material or an electron transporting material are not sufficient.

[Patent document 1]

U.S. Patent No. 4,539,507

5 [Patent document 2]

U.S. Patent No. 4,720,432

[Patent document 3]

U.S. Patent No. 4,885,211

[Patent document 4]

10 U.S. Patent No. 5,151,629

[Patent document 5]

U.S. Patent No. 5,409,783

[Patent document 6]

U.S. Patent No. 5,382,477

15 [Patent document 7]

U.S. Patent No. 5,130,603; U.S. Patent No.  
6,093,864

[Patent document 8]

U.S. Patent No. 5,227,252

20 [Patent document 9]

Japanese Patent Application Laid-Open No. H5-  
202356 (no corresponding foreign document)

[Patent document 10]

Japanese Patent Application Laid-Open No. H9-  
25 202878 (no corresponding foreign document)

[Patent document 11]

Japanese Patent Application Laid-Open No. H9-

227576 (no corresponding foreign document)

[Patent document 12]

U.S. Patent No. 5,247,190

[Patent document 13]

5 U.S. Patent No. 5,514,878

[Patent document 14]

U.S. Patent No. 5,672,678

[Patent document 15]

10 U.S. Patent No. 5,317,169; U.S. Patent No.  
5,726,457

[Patent document 16]

Japanese Patent Application Laid-Open No. H5-  
247460 (no corresponding foreign document)

[Patent document 17]

15 U.S. Patent No. 5,393,614

[Patent document 18]

Japanese Patent Application Laid-Open No. H7-  
82551 (no corresponding foreign document)

[Patent document 19]

20 U.S. Patent No. 6,010,796

[Patent document 20]

Japanese Patent Application Laid-Open No. 2001-  
267080 (no corresponding foreign document)

[Patent document 21]

25 Japanese Patent Application Laid-Open No. 2001-  
131174 (no corresponding foreign document)

[Non-patent document 1]

Appl. Phys. Lett. 51, 913 (1987)

[Non-patent document 2]

Nature, 395, 151 (1998)

[Non-patent document 3]

5 Nature, 347, 539 (1990)

#### DISCLOSURE OF THE INVENTION

It is, therefore, an object of the present invention to provide a novel phenanthroline compound.

10 It is another object of the present invention to provide an organic light emitting device having a light output with an extremely high efficiency and a high luminance using a specific phenanthroline compound.

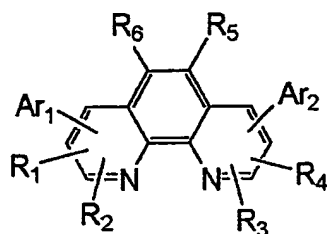
15 It is still another object of the present invention to provide an extremely durable organic light emitting device.

It is yet another object of the present invention to provide an organic light emitting device  
20 that is easily produced and can be prepared at a relatively low cost.

Specifically, the present invention provides a phenanthroline compound represented by any one of the following general formulas [I] to [III]:

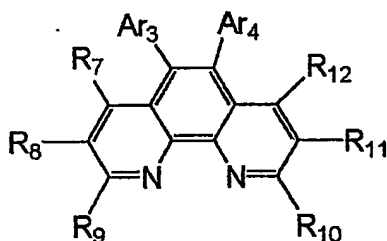
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[I]

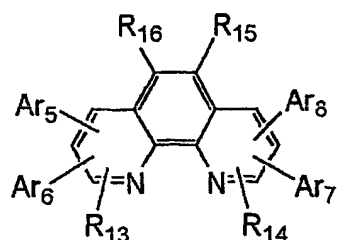
(wherein  $R_1$ ,  $R_2$ ,  $R_3$ ,  $R_4$ ,  $R_5$  and  $R_6$  are the same or different and each is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, and a halogen atom; and  $Ar_1$  and  $Ar_2$  are the same or different and each is selected from an unsubstituted or substituted fluorenyl group, an unsubstituted or substituted fluoranthenyl group, an unsubstituted or substituted perylenyl group, and an unsubstituted or substituted carbazolyl group);



[II]

(wherein  $R_7$ ,  $R_8$ ,  $R_9$ ,  $R_{10}$ ,  $R_{11}$  and  $R_{12}$  are the same or different and each is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, and

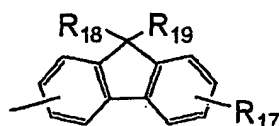
a halogen atom; and Ar<sub>3</sub> and Ar<sub>4</sub> are the same or different and each is selected from an unsubstituted or substituted fluorenyl group, an unsubstituted or substituted fluoranthenyl group, an unsubstituted or substituted perylenyl group, and an unsubstituted or substituted carbazolyl group); and



[III]

(wherein R<sub>13</sub>, R<sub>14</sub>, R<sub>15</sub> and R<sub>16</sub> are the same or different and each is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, and a halogen atom; and Ar<sub>5</sub>, Ar<sub>6</sub>, Ar<sub>7</sub> and Ar<sub>8</sub> are the same or different and each is selected from an unsubstituted or substituted fluorenyl group, an unsubstituted or substituted fluoranthenyl group, an unsubstituted or substituted perylenyl group, and an unsubstituted or substituted carbazolyl group).

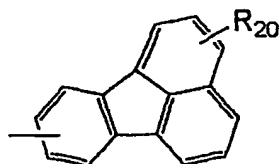
In the phenanthroline compounds of the present invention, the fluorenyl group is preferably represented by the following general formula [IV]:



[IV]

(wherein  $R_{17}$  is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, a substituted amino group, a cyano group, and a halogen atom; and  $R_{18}$  and  $R_{19}$  are the same or different and each is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, and an unsubstituted or substituted heterocyclic group).

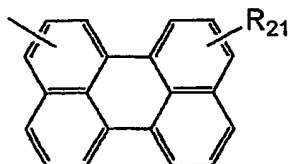
Also, the fluoranthenyl group is preferably represented by the following general formula [V]:



[V]

(wherein  $R_{20}$  is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, a substituted amino group, a cyano group, and a halogen atom).

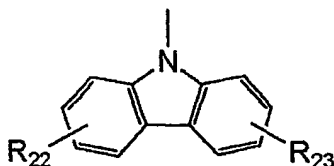
Also, the perylenyl group is preferably represented by the following general formula [VI]:



[VI]

(wherein  $R_{21}$  is selected from a hydrogen atom, an  
5 unsubstituted or substituted alkyl group, an  
unsubstituted or substituted aralkyl group, an  
unsubstituted or substituted aryl group, an  
unsubstituted or substituted heterocyclic group, a  
substituted amino group, a cyano group, and a halogen  
10 atom).

Also, the carbazolyl group is preferably represented by the following general formula [VII]:



[VII]

(wherein  $R_{22}$  and  $R_{23}$  are the same or different and  
15 each is selected from a hydrogen atom, an  
unsubstituted or substituted alkyl group, an  
unsubstituted or substituted aralkyl group, an  
unsubstituted or substituted aryl group, an  
unsubstituted or substituted heterocyclic group, a  
20 substituted amino group, a cyano group, and a halogen  
atom).

Further, the present invention provides an organic light emitting device comprising a pair of

electrodes consisting of an anode and a cathode, and a layer comprising an organic compound comprising at least one of the above-mentioned phenanthroline compounds, interposed between the pair of electrodes.

5

#### BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a sectional view illustrating an example of the organic light emitting device according to the present invention;

10 Fig. 2 is a sectional view illustrating another example of the organic light emitting device according to the present invention;

Fig. 3 is a sectional view illustrating still another example of the organic light emitting device according to the present invention;

15

Fig. 4 is a sectional view illustrating yet another example of the organic light emitting device according to the present invention;

Fig. 5 is a sectional view illustrating yet still another example of the organic light emitting device according to the present invention; and

20

Fig. 6 is a sectional view illustrating yet again another example of the organic light emitting device according to the present invention.

25

#### BEST MODE FOR CARRYING OUT THE INVENTION

The present invention will now be described in

detail.

The phenanthroline compounds of the present invention will be first described.

Preferably, the phenanthroline compounds of the present invention are represented by the above  
5 general formulas [I] to [III], wherein a fluorenyl group is represented by the above general formula [IV], a fluoranthenyl group by the above general formula [V], a perylenyl group by the above general  
10 formula [VI] and a carbazolyl group by the above general formula [VII].

Specific examples for the substituent groups in the above general formulas [I]-[VII] are shown below.

The alkyl group includes methyl, ethyl, n-  
15 propyl, iso-propyl, n-butyl, ter-butyl, octyl or the like.

The aralkyl group includes benzyl, phenethyl or the like.

The aryl group includes phenyl, biphenyl,  
20 terphenyl or the like.

The heterocyclic group includes thienyl, pyrrolyl, pyridyl, oxazolyl, oxadiazolyl, thiazolyl, thidiazolyl, terthienyl or the like.

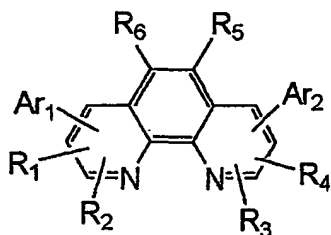
The substituted amino group includes  
25 dimethylamino, diethylamino, dibenzylamino, diphenylamino, ditolylamino, dianisolylamino or the like.

- 13 -

The halogen atom includes fluorine, chlorine, bromine, iodine or the like.

The substituent groups that the above substituent groups may have include alkyl groups such as methyl, ethyl and propyl; aralkyl groups such as benzyl and phenethyl; aryl groups such as phenyl and biphenyl; heterocyclic groups such as thienyl, pyrrolyl and pyridyl; amino groups such as dimethylamino, diethylamino, dibenzylamino, diphenylamino, ditolylamino and dianisolyamino; alkoxy groups such as methoxyl, ethoxyl, propoxyl and phenoxy; cyano group; and halogen atoms such as fluorine, chlorine, bromine and iodine.

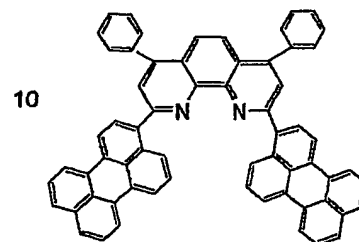
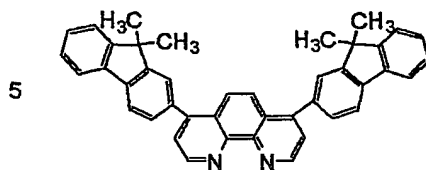
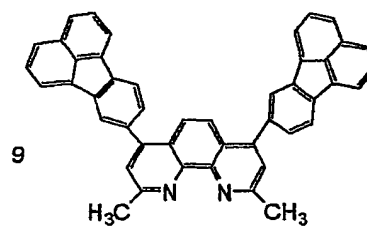
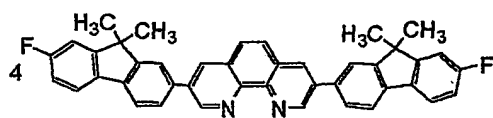
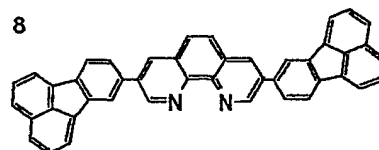
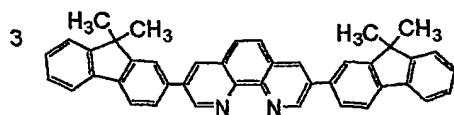
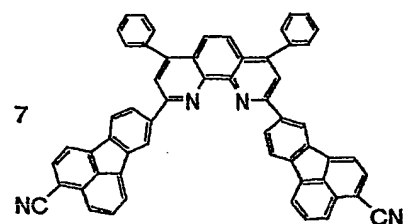
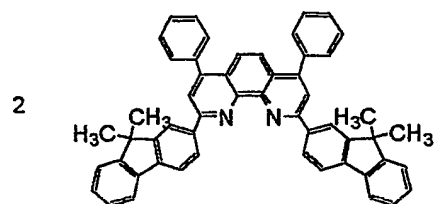
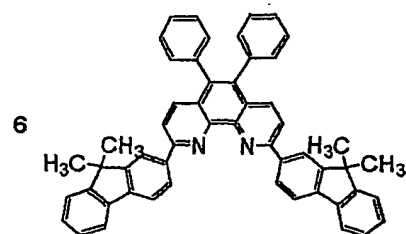
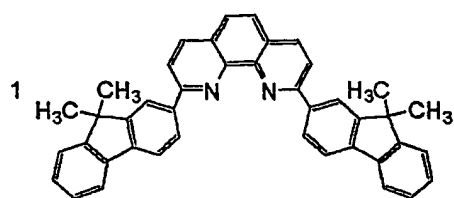
The followings are typical examples of the phenanthroline compounds of the present invention, but the present invention is not limited thereto:



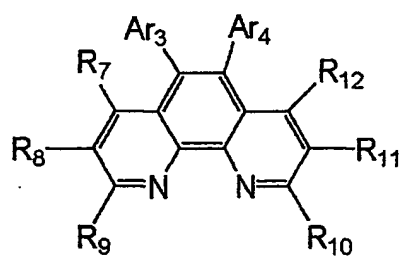
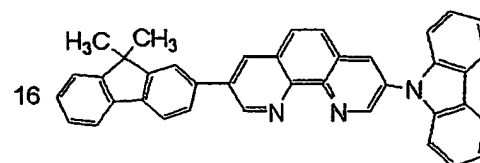
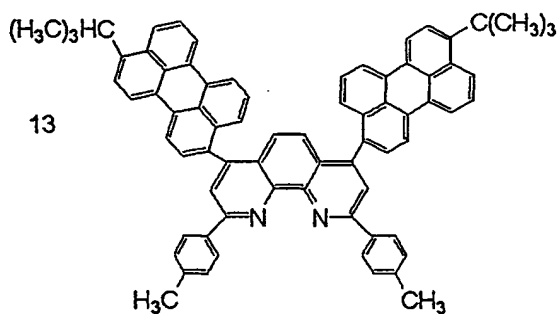
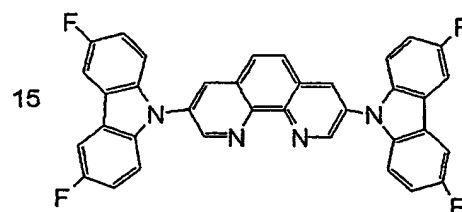
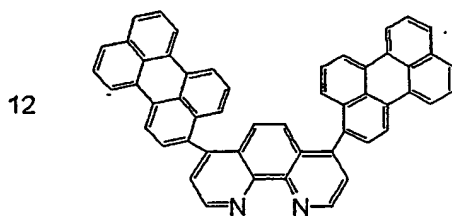
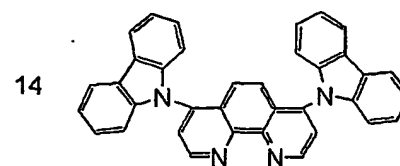
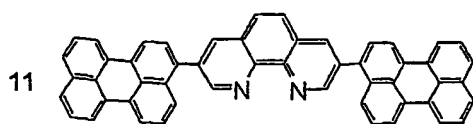
[I]

[Typical Examples of Compounds of Formula [I]]

(Exemplary Compound Nos. 1-16)



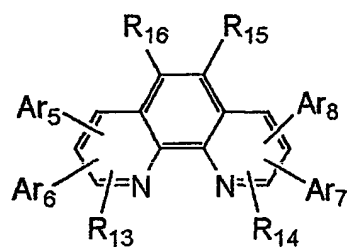
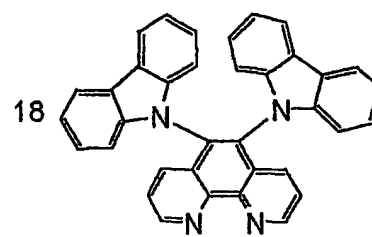
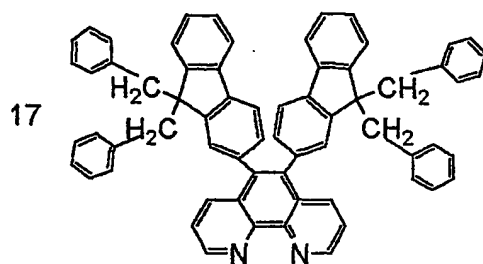




[II]

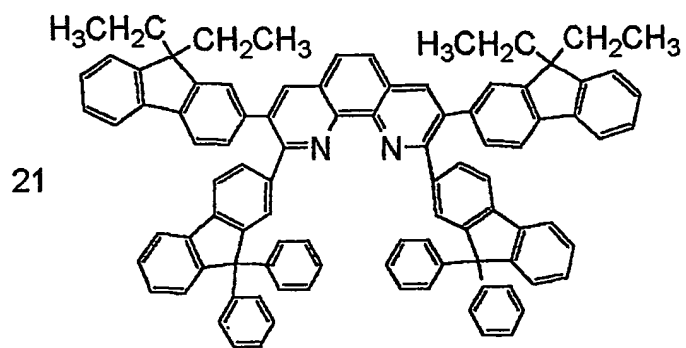
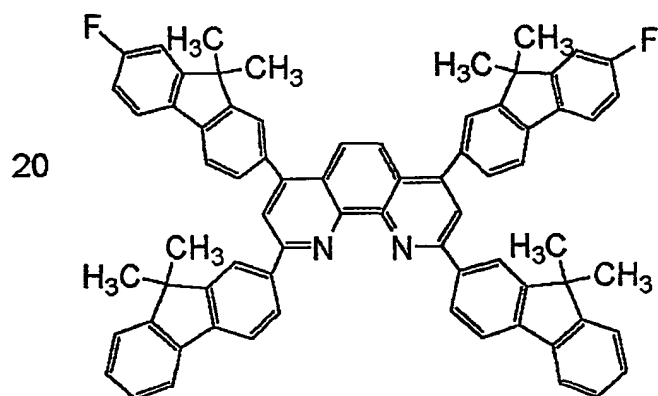
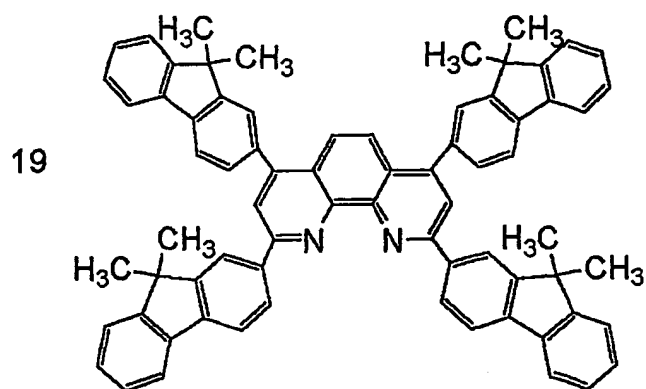
[Typical Examples of Compounds of Formula [II]]

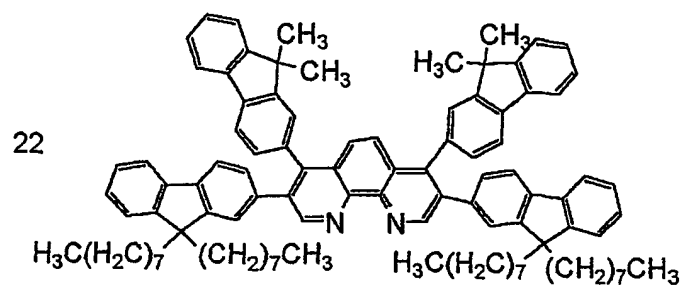
5 (Exemplary Compound Nos. 17-18)



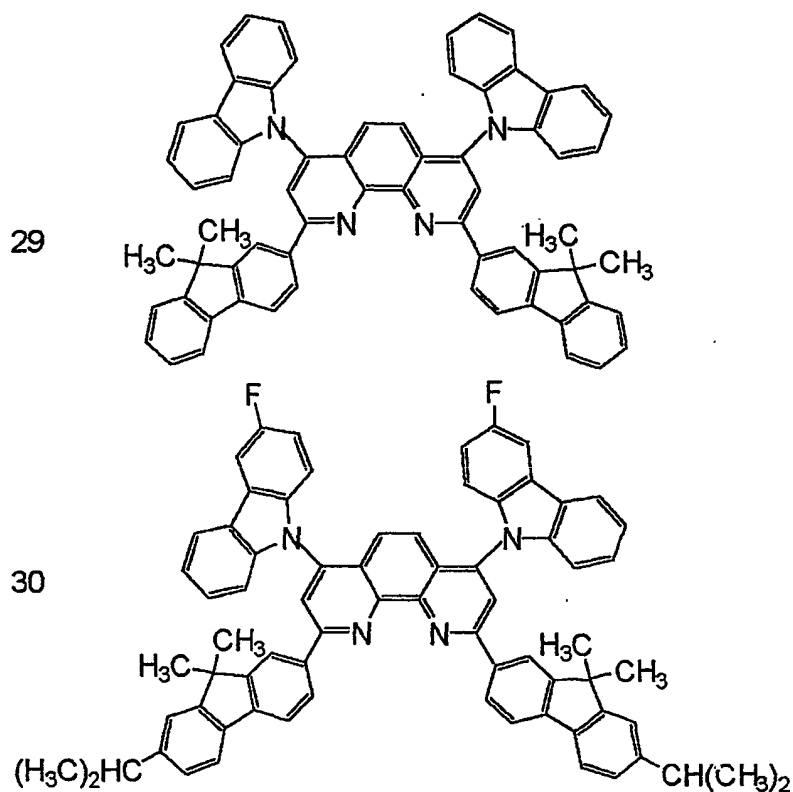
[III]

[Typical Examples of Compounds of Formula [III]]  
(Exemplary Compound Nos. 19-30)









The phenanthroline compound of the present invention can be synthesized by generally known methods, in which it can be obtained by the synthesis  
5 such as the Suzuki coupling method using a palladium catalyst (e.g., Chem. Rev. 1995, 95, 2457-2483) through a phenanthroline compound intermediate that is obtained by the methods, for example, described in J. Org, Chem., 16, 941-945 (1951); Tetrahedron, Lett.,  
10 36, 3489-3490 (1995) and the like.

The phenanthroline compound of the present invention has superior electron transporting property and durability to the conventional compounds and is useful as a layer comprising an organic compound of

an organic light emitting device, particularly as an electron transporting layer and a light emitting layer. Moreover, the layers formed by a vacuum evaporation process or a solution coating process are  
5 difficult to be crystallized and are excellent in long-term stability.

The organic light emitting device of the present invention will now be described in detail.

The organic light emitting device of the  
10 present invention comprises a pair of electrodes consisting of an anode and a cathode, and a layer comprising an organic compound comprising at least one of the phenanthroline compounds represented by the general formula [I], [II] and [III], interposed  
15 between the pair of electrodes.

In the organic light emitting device of the present invention, it is preferred that at least an electron transporting layer or a light emitting layer of the layer(s) comprised of an organic compound  
20 comprises at least one of the above-mentioned phenanthrolines.

In the organic light emitting device of the present invention, the phenanthroline compound represented by the above general formulas [I] to  
25 [III] is formed between the anode and the cathode by a vacuum evaporation process or a solution coating process. The organic layer is preferably formed in a

thin film having a thickness of less than 10  $\mu\text{m}$ , preferably 0.5  $\mu\text{m}$  or less, more preferably from 0.01 to 0.5  $\mu\text{m}$ .

Figs. 1 to 6 are views illustrating preferred  
5 examples of the organic light emitting device of the present invention.

Fig. 1 is a sectional view illustrating one example of the organic light emitting device of the present invention. The device of Fig. 1 has the  
10 structure in which an anode 2, a light emitting layer 3 and a cathode 4 are provided on a substrate 1 in the mentioned order. The structure shown in Fig. 1 is useful when employing a compound having hole transportability, electron transportability and light  
15 emitting property singularly within itself, or when employing compounds having respective characteristics in mixture.

Fig. 2 is a sectional view illustrating another example of the organic light emitting device of the present invention. The device of Fig. 2 has the  
20 structure in which an anode 2, a hole transporting layer 5, an electron transporting layer 6 and a cathode 4 are provided on a substrate 1 in the mentioned order. The structure shown in Fig. 2 is  
25 useful when a material having a hole transportability and/or electron transportability is used for respective layers as a light emitting substance in



combination with a hole transporting or electron transporting substance having no light emitting property. In this case, the light emitting layer comprises either the hole transporting layer 5 or the  
5 electron transporting layer 6.

Fig. 3 is a sectional view illustrating another example of the organic light emitting device of the present invention. The device of Fig. 3 has the structure in which an anode 2, a hole transporting  
10 layer 5, a light emitting layer 3, an electron transporting layer 6 and a cathode 4 are provided on a substrate 1 in the mentioned order. The structure is to separate a carrier transporting function and a light emitting function, and is used in suitable  
15 combination with compounds having the respective properties of hole transporting property, electron transporting property and light emitting property. Thus, the freedom of selection of materials is extremely increased, and various compounds having  
20 different emission wavelengths can be used to allow diversification of the luminescent hue. Further, carriers or excitons can be effectively confined in the central light emitting layer 3 to improve the light emission efficiency.

25 Fig. 4 is a sectional view illustrating another example of the organic light emitting device of the present invention. The device of Fig. 4 has the

- 24 -

structure in which a hole injecting layer 7 is inserted to the anode 2 side as compared with the structure shown in Fig. 3. The structure shown in Fig. 1 is effective for improving adhesiveness of the anode 2 to the hole transporting layer 5 or improving the hole injecting property and is also effective for driving at a reduced voltage.

Figs. 5 and 6 are sectional views illustrating other examples of the organic light emitting device of the present invention. The devices of Figs. 5 and 6 have the structures in which a layer (hole-blocking layer 8) for blocking holes or excitons from passing through to the cathode 4 side is inserted between the light emitting layer 3 and the electron transporting layer 6 as compared with Figs. 3 and 4. The structure is effective for improving the light emission efficiency by using a compound having a very high ionization potential as the hole blocking layer 8.

It should be noted that Figs. 1 to 6 merely show very basic device structures, and the structures of the organic light emitting device using the compounds of the present invention are not limited thereto. It is possible to take various structures, for example, to provide an insulating layer at an interface between an electrode and an organic layer, to provide an adhesion layer or an interference layer

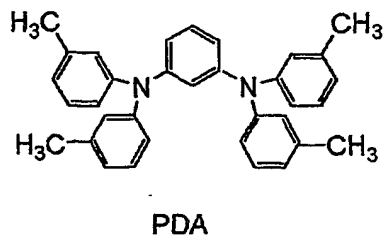
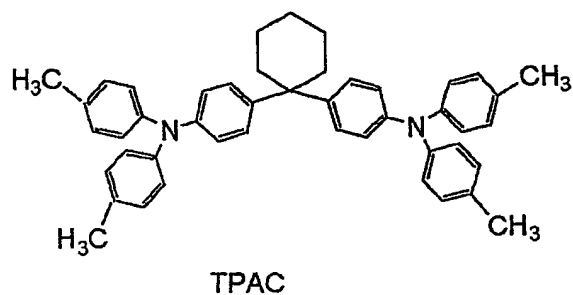
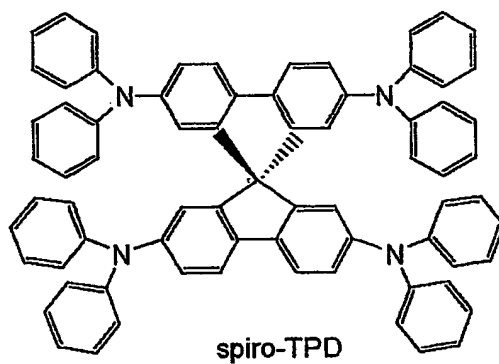
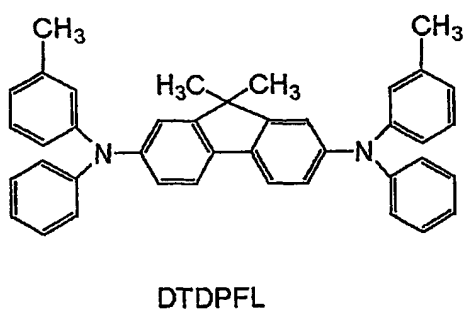
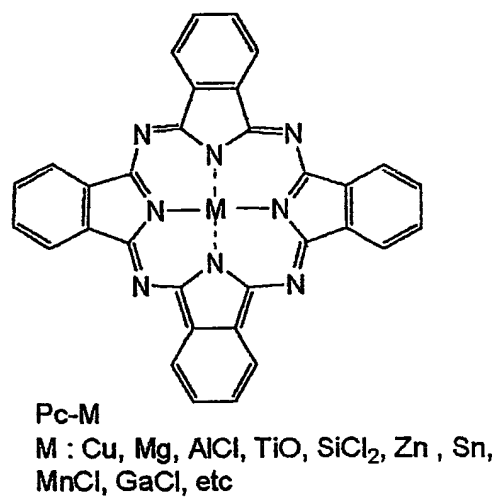
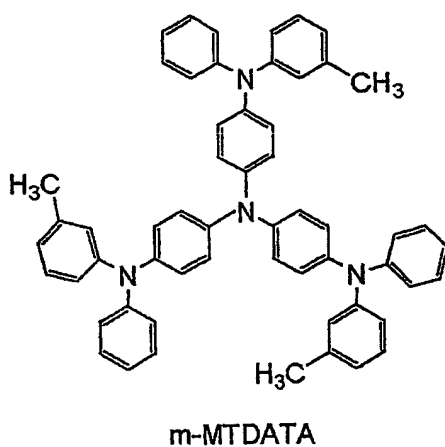
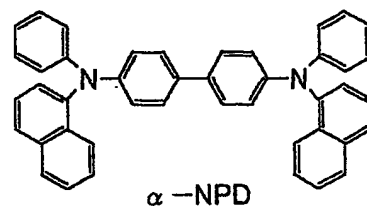
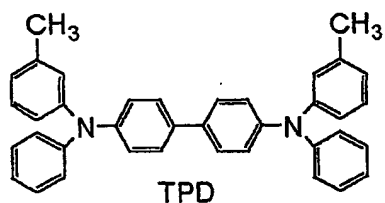
or to compose a hole transporting layer of two layers having different ionization potentials.

The phenanthroline compounds represented by the general formulas [I] to [III] used in the present  
5 invention are excellent in electron transporting property and durability compared with the conventional compounds, and can be used in any one of the structures shown in Figs. 1 to 6.

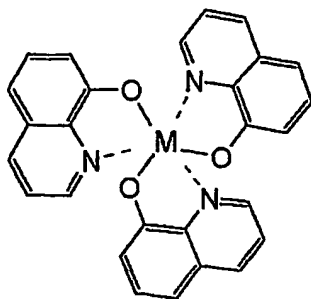
Although the present invention uses the  
10 phenanthroline compounds represented by the general formulas [I] to [III] as constituent components for an electron transporting layer or a light emitting layer, already known hole transporting compounds, light emitting compounds or electron transporting  
15 compounds can also be used together as needed

Examples of these compounds include the followings:

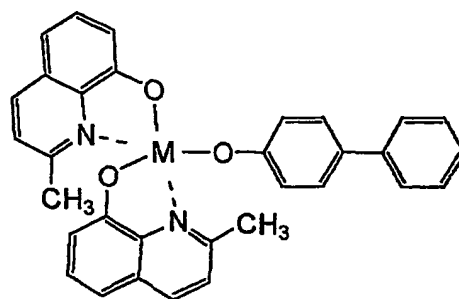
## [Hole Transporting Compounds]



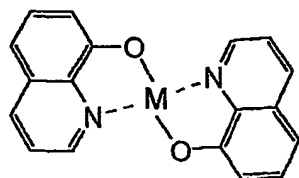
## [Electron Transporting, Light Emitting Compounds]



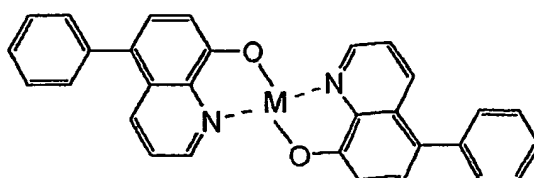
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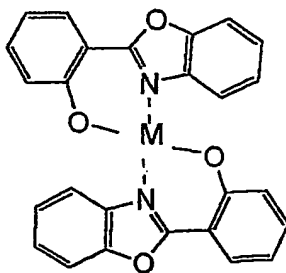
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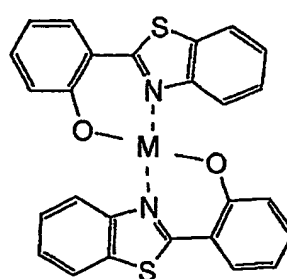
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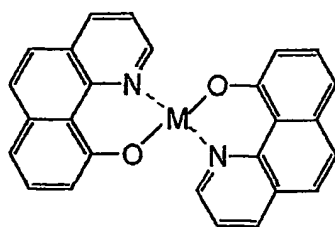
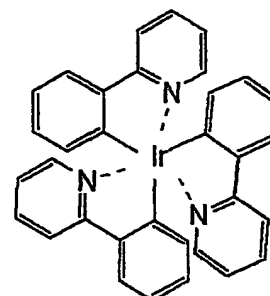
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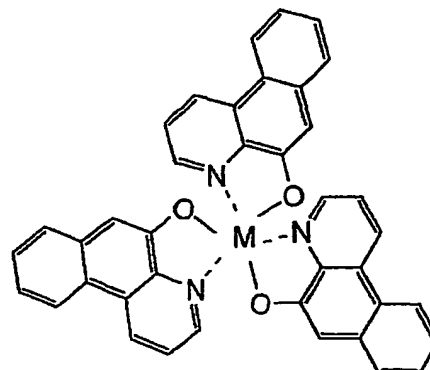
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M : Zn , Mg , Be

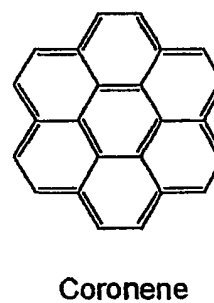
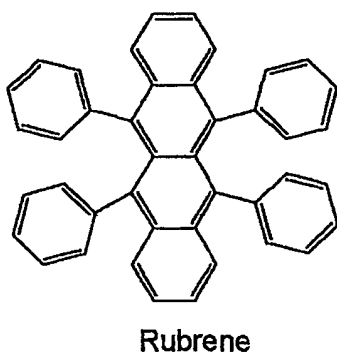
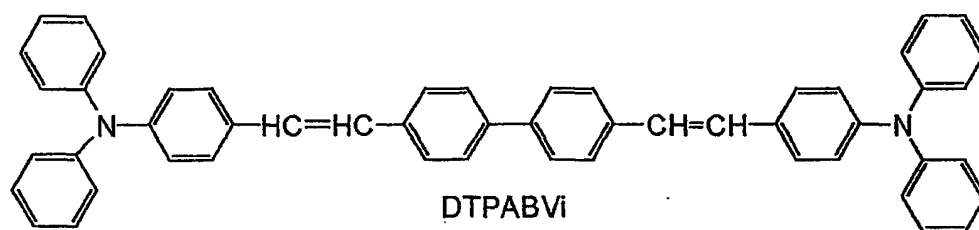
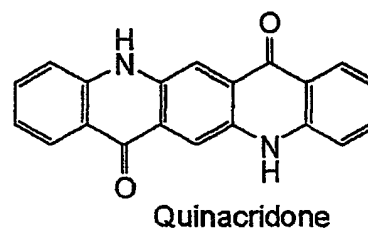
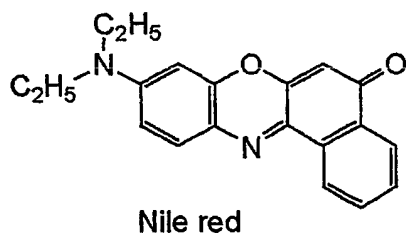
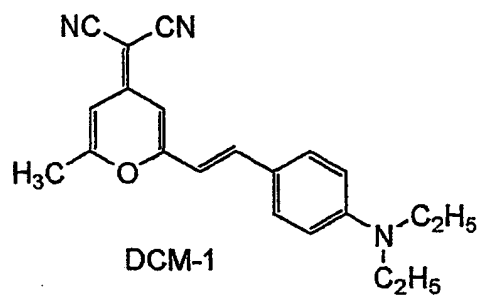
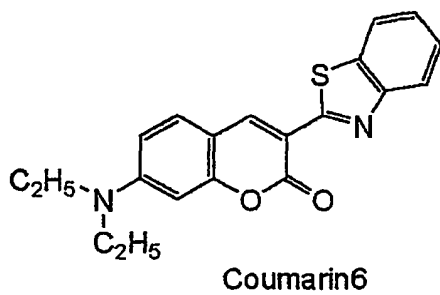


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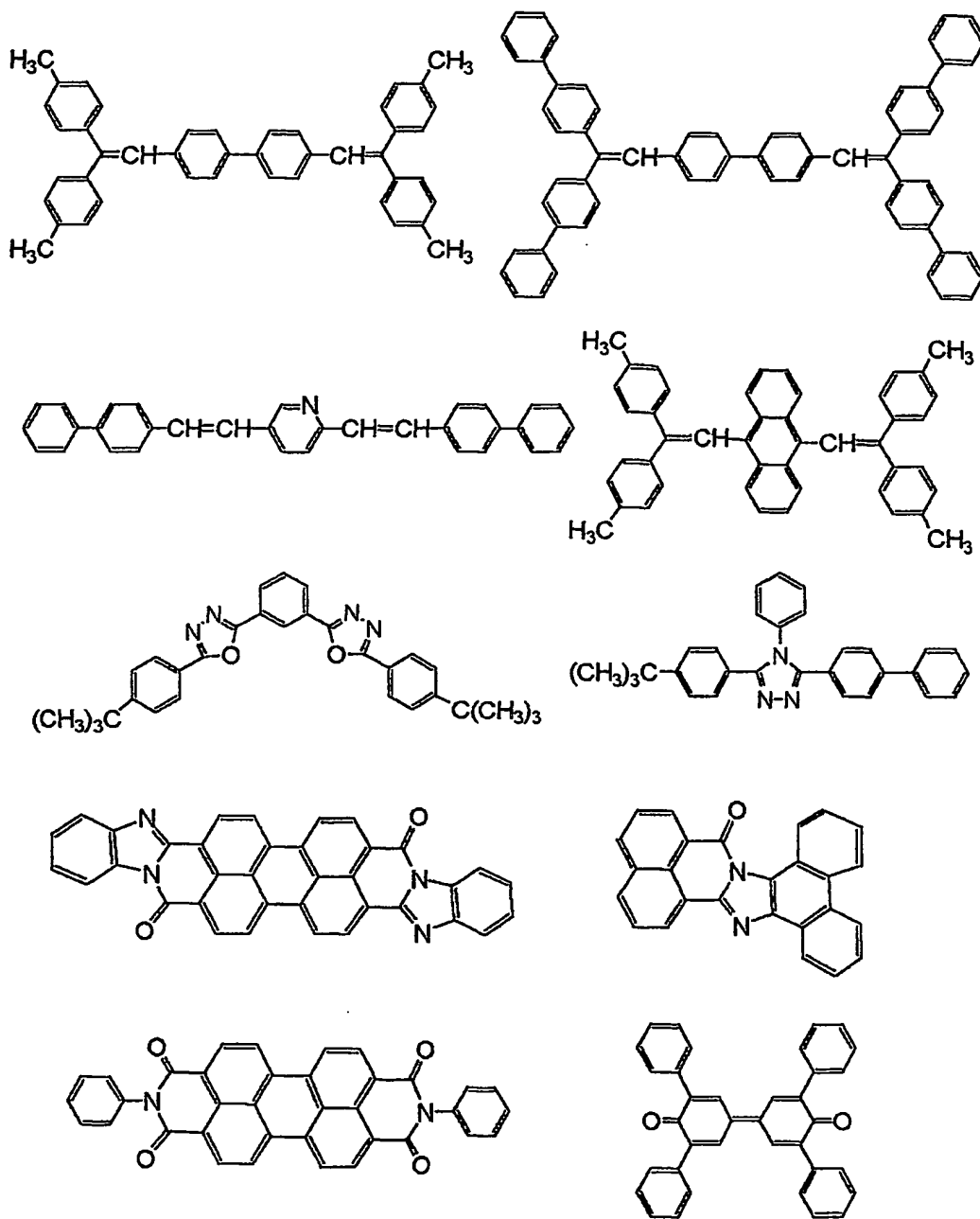


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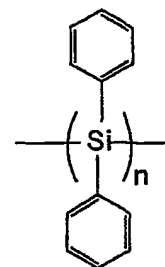
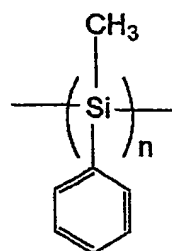
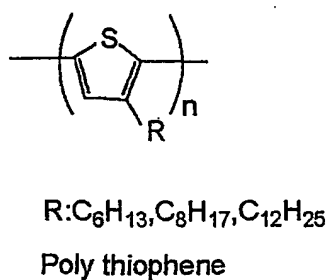
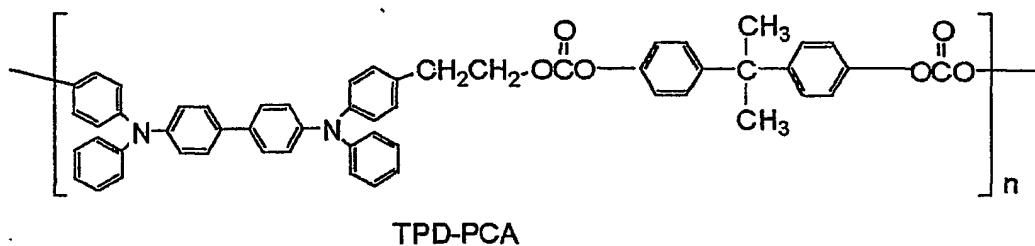
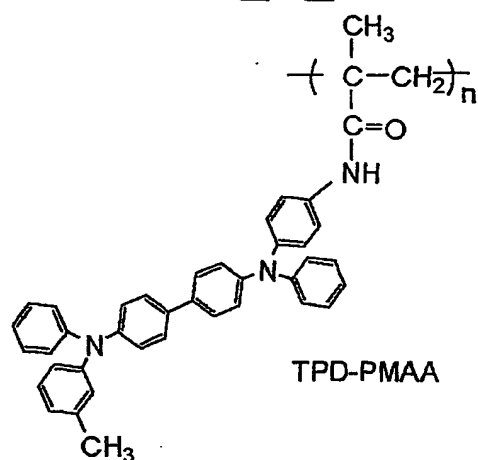
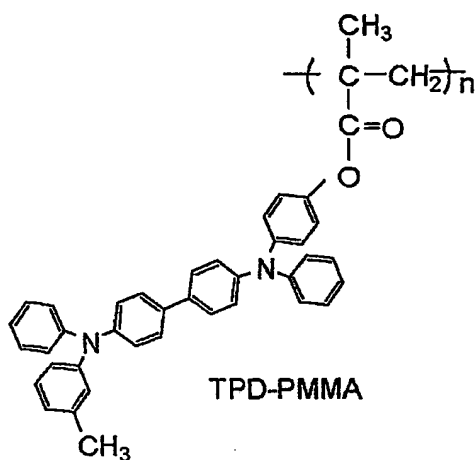
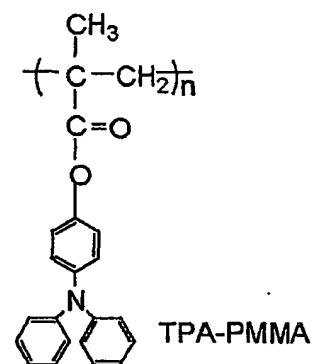
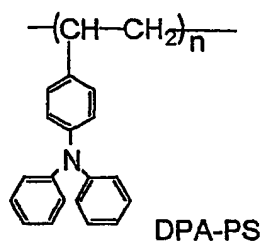
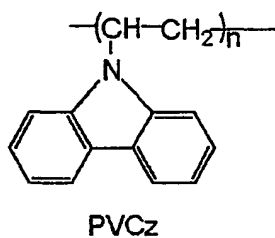
## [Light Emitting Compounds]



[Light Emitting Layer Matrix Compounds/Electron  
Transporting Compounds]

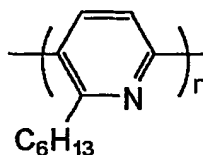
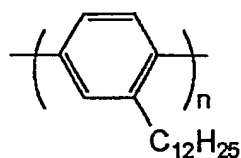
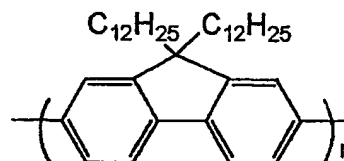
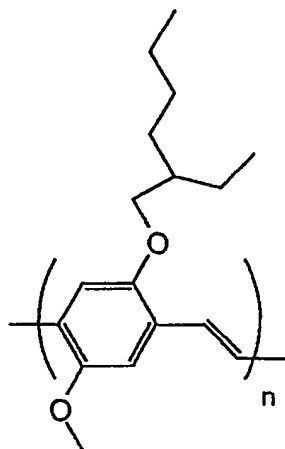
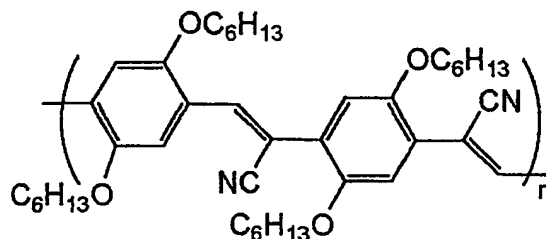
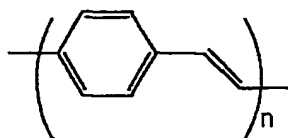


## [Polymer-based Hole Transporting Compounds]





[Polymer-based Light Emitting  
Compounds/Charge Transporting Compounds]



In the organic light emitting device of the present invention, the layers containing the phenanthroline compounds represented by the general formulas [I] to [III] and the layers containing other organic compounds are generally formed into thin films by a vacuum evaporation process or a coating process in which they are dissolved in a suitable solvent. In particular, when the film is formed by a coating process, it is also possible to form the film in combination with a suitable binder resin.

The binder resin can be selected from a wide range of binder resins, and include, for example, but not limited to, polyvinylcarbazole resins, polycarbonate resins, polyester resins, polyarylate resins, polystyrene resins, acrylic resins, methacrylic resins, butyral resins, polyvinylacetal resins, diallylphthalate resins, phenol resins, epoxy resins, silicone resins, polysulfone resins, urea resins and the like. In addition, one of them or a mixture of two or more of them may be used in the form of a homopolymer or a copolymer.

The materials for the anode preferably have a large work function, and metals such as, for example, gold, platinum, nickel, palladium, cobalt, selenium, vanadium and alloys thereof and metal oxides such as tin oxides, zinc oxides, indium tin oxides (ITO) and indium zinc oxides can be used. In addition, conductive polymers such as polyaniline, polypyrrole, polythiophene and polyphenylene sulfide can be used. These electrode materials can be used singularly or in combination.

On the other hand, the materials for the cathode preferably have a small work function, and metals such as lithium, sodium, potassium, cesium, calcium, magnesium, aluminium, indium, silver, lead, tin and chrome and alloys thereof can be used. Metal oxides such as indium tin oxides (ITO) can also be

- 33 -

used. Moreover, the cathode may have either a one-layer structure or a multilayer structure.

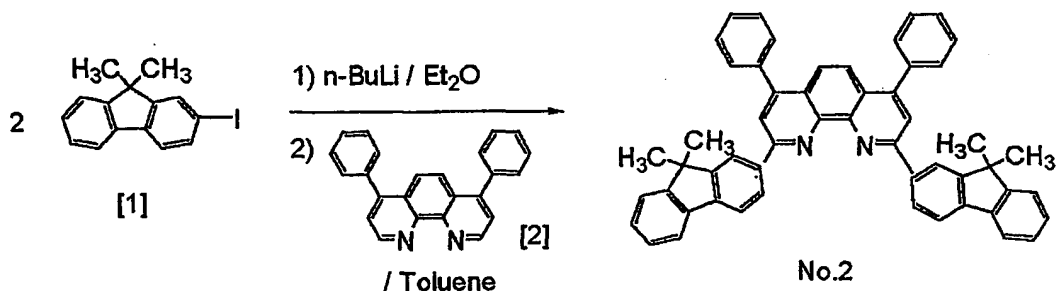
The substrate for use in the present invention includes, but not limited to, metal substrates, opaque substrates such as ceramic substrates, and transparent substrates such as glass, quartz and plastic sheets. Moreover, it is possible to control the color of emitted light using a color filter film, a fluorescent color conversion filter film, a dielectric reflecting film and the like for the substrate.

Furthermore, a protective layer or an encapsulant layer can also be provided on the prepared device for the purpose of preventing contact with oxygen, moisture and the like. The protective layer includes an inorganic material film such as a diamond thin film, a metal oxide film or a metal nitride film; a polymeric film such as of fluoro-resin, polyparaxylene, polyethylene, silicone resin and polystyrene resin; a photo-curable resin or the like. Moreover, the device itself can be covered with glass, a gas-impermeable film, metal or the like and packaged with a suitable encapsulant resin.

#### [Examples]

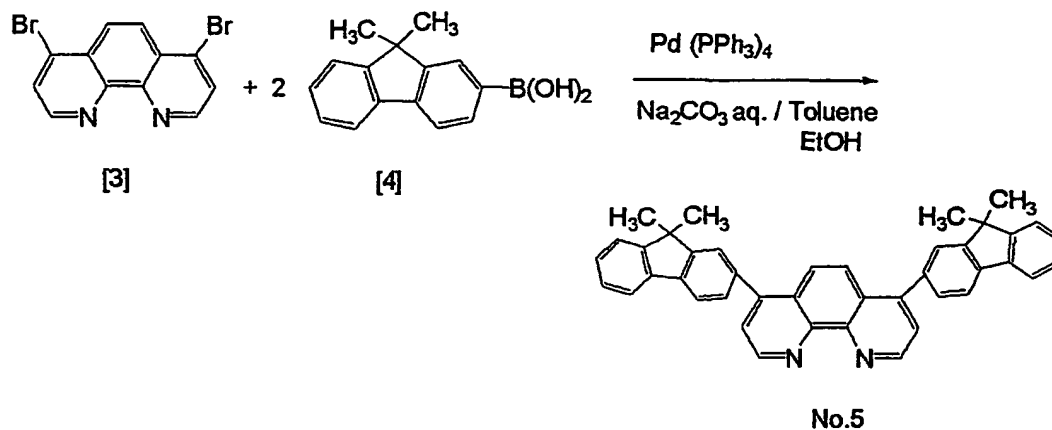
The present invention will now be described in detail with reference to examples, but the present invention is not limited to them.

[Synthesis Example 1] [Synthesis of Exemplary Compound No. 2]



- To a three-necked flask of 300 ml, 5.8 g (18.1 mmol) of 2-iodo-9,9-dimethylfluorene [1] and 80 ml of diethyl ether were charged and 11.7 ml (18.1 mmol) of n-butyllithium (hexane solution of 15%) was dropped under stirring at -78°C in a nitrogen atmosphere. The mixture was raised to room temperature and stirred for one hour, and then cooled to -20°C and a dispersion of 1.5 g (4.51 mmol) of bathophenanthroline [2] in 100 ml of toluene was dropped. The mixture was stirred at room temperature for 12 hours and was added with water. The organic layer was extracted with chloroform and dried with anhydrous sodium sulfate, and then purified with an alumina column (hexane/chloroform solvent mixture developer), obtaining 2.4 g (yield of 74%) of Exemplary Compound No. 2 (yellow crystal).
- [Synthesis Example 2] [Synthesis of Exemplary Compound No. 5]

- 35 -



\*1) J.Org.Chem., 16, 941-945 (1951)

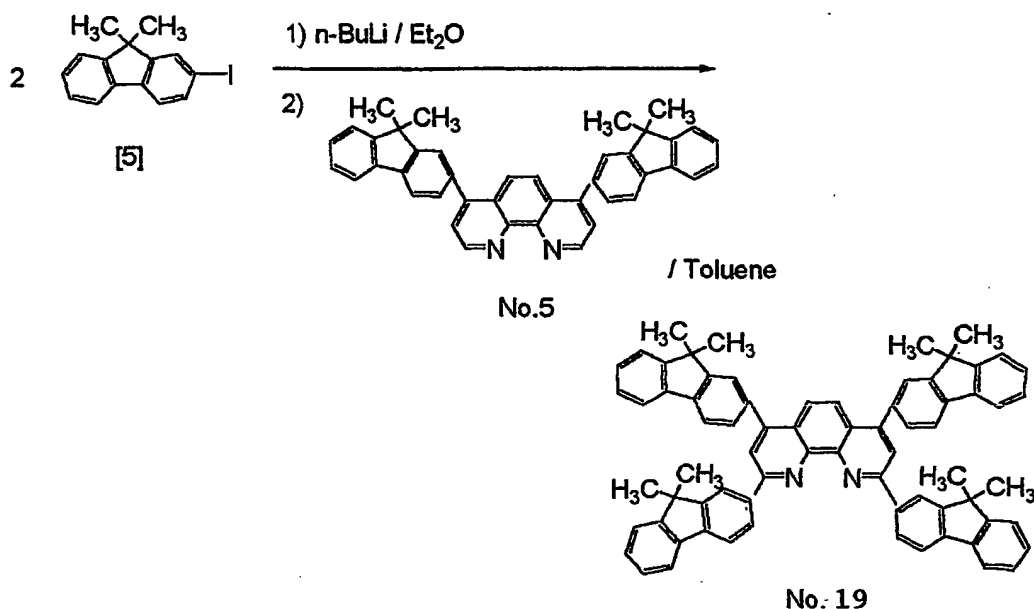
To a three-necked flask of 500 ml, 1.0 g (2.96 mmol) of 4,7-dibromo-1,10-phenanthroline [3]<sup>\*1)</sup>, 2.8 g (11.8 mmol) of 9,9-dimethylfluorene-2-boronic acid [4], 140 ml of toluene and 70 ml of ethanol were charged and an aqueous solution of 12 g of sodium carbonate/60 ml of water was dropped under stirring at room temperature in a nitrogen atmosphere, and then 0.17 g (0.15 mmol) of

10 tetrakis(triphenylphosphine)palladium (0) was added. After stirring at room temperature for 30 minutes, the mixture was raised to a temperature of 77°C and stirred for 3 hours. After the reaction, the organic layer was extracted with chloroform and dried with

15 anhydrous sodium sulfate, and then purified with an alumina column (hexane/chloroform solvent mixture developer), obtaining 1.5 g (yield of 90%) of Exemplary Compound No. 5 (white crystal).

[Synthesis Example 3] [Synthesis of Exemplary

Compound No. 19]

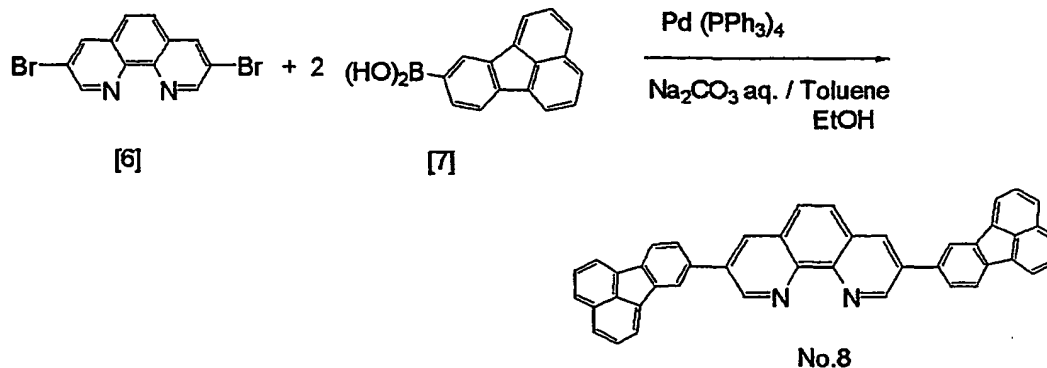


To a three-necked flask of 300 ml, 2.3 g (7.18 mmol) of 2-iodo-9,9-dimethylfluorene [5] and 60 ml of diethyl ether were charged and 4.6 ml (7.18 mmol) of  $n$ -butyllithium (hexane solution of 15%) was dropped under stirring at  $-78^\circ\text{C}$  in a nitrogen atmosphere. The mixture was raised to room temperature and stirred for one hour, and then cooled to  $-20^\circ\text{C}$  and a dispersion of 1.0 g (1.77 mmol) of Exemplary Compound No. 5 in 80 ml of toluene was dropped. The mixture was stirred at room temperature for 12 hours and was added with water. The organic layer was extracted with chloroform and dried with anhydrous sodium sulfate, and then purified with an alumina column (hexane/chloroform solvent mixture developer), obtaining 1.2 g (yield of 73%) of Exemplary Compound

- 37 -

No. 19 (yellow crystal).

[Synthesis Example 4] [Synthesis of Exemplary Compound No. 8]



\*2) Tetrahedron, Lett., 36, 3489-3490 (1995)

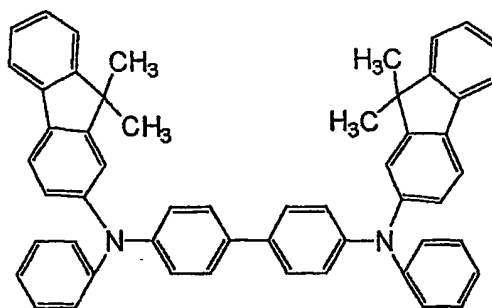
- 5            To a three-necked flask of 500 ml, 1.0 g (2.96 mmol) of 3,8-dibromo-1,10-phenanthroline [6]<sup>\*2)</sup>, 2.9 g (11.8 mmol) of fluorantene-8-boronic acid [7], 140 ml of toluene and 70 ml of ethanol were charged and an aqueous solution of 12 g of sodium carbonate/60 ml of
- 10    water was dropped under stirring at room temperature in a nitrogen atmosphere, and then 0.17 g (0.15 mmol) of tetrakis(triphenylphosphine)palladium (0) was added. After stirring at room temperature for 30 minutes, the mixture was raised to a temperature of
- 15    77°C and stirred for 3 hours. After the reaction, the organic layer was extracted with chloroform and dried with anhydrous sodium sulfate, and then purified with an alumina column (hexane/chloroform solvent mixture developer), obtaining 1.4 g (yield of
- 20    82%) of Exemplary Compound No. 8 (yellow crystal).

## (Example 1)

A device having the structure shown in Fig. 3 was prepared.

On a glass substrate as the substrate 1, indium  
5 tin oxide (ITO) as the anode 2 was deposited by a sputtering process in a thickness of 120 nm, the resultant structure being used as a transparent conductive supporting substrate. This was  
10 ultrasonically cleaned with acetone and isopropyl alcohol (IPA) in this order, and dried after the cleaning by boiling with IPA. Further, it was cleaned with UV/ozone. The resultant structure was used as a transparent conductive supporting substrate.

On the transparent conductive supporting  
15 substrate, a chloroform solution of the compound represented by the following structural formula was applied by a spin-coating process to form a film having a thickness of 30 nm, thus forming the hole transporting layer 5.

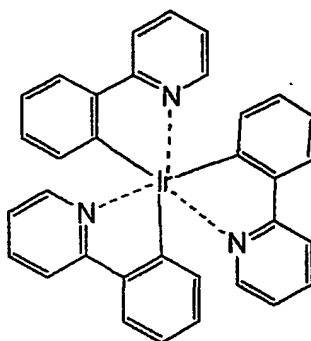


20

In addition, the Ir complex represented by the following structural formula and Exemplary Compound



No. 1 as the instant phenanthroline compound (weight ratio of 5:100) were deposited by a vacuum evaporation process in a thickness of 20 nm to form the light emitting layer 3. As for the conditions, the degree of the vacuum at the evaporation was  $1.0 \times 10^{-4}$  Pa and the film formation rate was 0.2-0.3 nm/sec.



Moreover, trisquinolinol aluminium was deposited by a vacuum evaporation process in a thickness of 40 nm to form the electron transporting layer 6. As for the conditions, the degree of the vacuum at the evaporation was  $1.0 \times 10^{-4}$  Pa and the film formation rate was 0.2-0.3 nm/sec.

Then, as the cathode 4, a vacuum evaporation material consisting of aluminium and lithium (lithium concentration of 1 atomic %) was used to form a metal layer film having a thickness of 50 nm on the above organic layer by a vacuum evaporation process, and further by the vacuum evaporation process an aluminium layer having a thickness of 150 nm was formed. As for the conditions, the degree of the vacuum at the evaporation was  $1.0 \times 10^{-4}$  Pa and the

film formation rate was 1.0-1.2 nm/sec.

Furthermore, the resultant structure was covered with a protective glass plate in a nitrogen atmosphere and sealed with an acrylic resin adhesive.

5        When the thus obtained device was applied with a DC voltage of 10 V using the ITO electrode (anode 2) as a positive electrode and the Al-Li electrode (cathode 4) as a negative electrode, the current passed through the device at a current density of  
10 18.0 mA/cm<sup>2</sup> and emission of green light was observed at a luminance of 4,500 cd/m<sup>2</sup>.

In addition, when the voltage was applied for 100 hours while maintaining the current density at 6.0 mA/cm<sup>2</sup>, the initial luminance of 850 cd/m<sup>2</sup> dropped  
15 to 800 cd/m<sup>2</sup> after 100 hours, exhibiting only a small reduction of luminance.

(Examples 2 to 9)

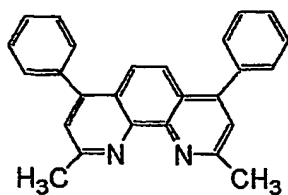
Devices were prepared and evaluated in the same manner as in Example 1 with the exception that  
20 Exemplary Compounds shown in Table 1 below were used instead of Exemplary Compound No. 1. The results are shown in Table 1.

(Comparative Examples 1 to 3)

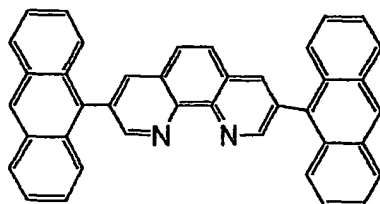
Devices were prepared and evaluated in the same  
25 manner as in Example 1 with the exception that Comparative Compound Nos. 1-3 shown below were used instead of Exemplary Compound No. 1. The results are

shown in Table 1.

Comparative Compound No. 1



Comparative Compound No. 2



Comparative Compound No. 3

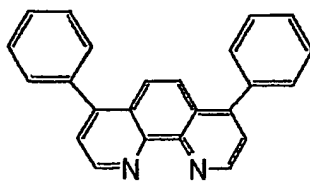


Table 1

Example No.	Exemplary Compound No.	Initial		Current Density (mA/cm <sup>2</sup> )	Durability	
		Applied Voltage (V)	Luminance (cd/m <sup>2</sup> )		Initial Luminance (cd/m <sup>2</sup> )	Luminance after 100 hours (cd/m <sup>2</sup> )
Example 1	1	10	4500	6.0	850	800
Example 2	2	10	5000	6.0	1040	900
Example 3	9	10	4300	6.0	760	680
Example 4	11	10	4100	6.0	760	590
Example 5	16	10	4320	6.0	800	590
Example 6	19	10	4900	6.0	1000	750
Example 7	22	10	4530	6.0	900	745
Example 8	27	10	4400	6.0	830	670
Example 9	30	10	4600	6.0	880	750
Comparative Example 1	Comparative Compound No. 1	10	760	6.0	430	200
Comparative Example 2	Comparative Compound No. 2	10	400	6.0	280	140
Comparative Example 3	Comparative Compound No. 3	10	1200	6.0	730	300

(Example 10)

A device of the structure shown in Fig. 3 was prepared.

The hole transporting layer 5 was formed on the transparent conductive supporting substrate in the same manner as in Example 1.

In addition, coumarin and trisquinolinol aluminium (polymerization ratio of 1:20) were deposited by a vacuum evaporation process in a thickness of 20 nm to form the light emitting layer 3. As for the conditions, the degree of the vacuum at the evaporation was  $1.0 \times 10^{-4}$  Pa and the film formation rate was 0.2-0.3 nm/sec.

Moreover, Exemplary Compound No. 3 was deposited in a thickness of 40 nm to form the electron transporting layer 6. As for the conditions, the degree of the vacuum at the evaporation was  $1.0 \times 10^{-4}$  Pa and the film formation rate was 0.2-0.3 nm/sec.

Then, the device was sealed after the cathode 4 was formed in the same manner as in Example 1.

When the thus obtained device was applied with a DC voltage of 8 V using the ITO electrode (anode 2) as a positive electrode and the Al-Li electrode (cathode 4) as a negative electrode, the current passed through the device at a current density of 1,110 mA/cm<sup>2</sup> and emission of green light was observed at a luminance of 95,000 cd/m<sup>2</sup>.

Furthermore, when the voltage was applied for 100 hours while maintaining the current density at 200 mA/cm<sup>2</sup>, the initial luminance of 10,000 cd/m<sup>2</sup> dropped to 8,500 cd/m<sup>2</sup> after 100 hours, exhibiting only a small reduction of luminance.

(Examples 11 to 18)

Devices were prepared and evaluated in the same manner as in Example 10 with the exception that Exemplary Compounds shown in Table 2 were used instead of Exemplary Compound No. 3. The results are shown in Table 2.

(Comparative Examples 4 to 6)

Devices were prepared and evaluated in the same manner as in Example 10 with the exception that Comparative Compound Nos. 1 to 3 were used instead of Exemplary Compound No. 3. The results are shown in Table 2.

Table 2

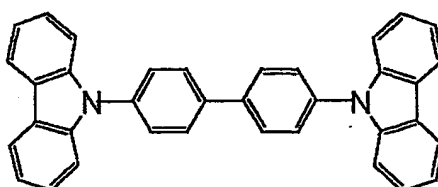
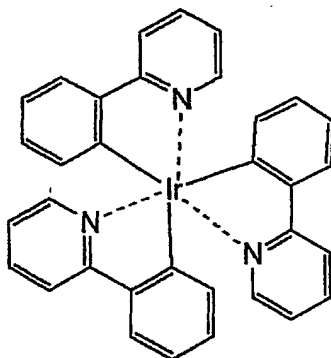
Example No.	Exemplary Compound No.	Initial		Durability		
		Applied Voltage (V)	Luminance (cd/m <sup>2</sup> )	Current Density (mA/cm <sup>2</sup> )	Initial Luminance (cd/m <sup>2</sup> )	Luminance after 100 hours (cd/m <sup>2</sup> )
Example 10	3	8	95000	200	10000	8500
Example 11	1	8	96000	200	14000	8750
Example 12	8	8	83000	200	13800	8680
Example 13	10	8	80000	200	13050	8700
Example 14	15	8	96000	200	13000	8540
Example 15	21	8	98000	200	15000	9750
Example 16	23	8	79000	200	11500	8600
Example 17	26	8	78000	200	11000	8560
Example 18	28	8	84000	200	13000	8800
Comparative Example 4	Comparative Compound No. 1	8	14000	200	7000	3500
Comparative Example 5	Comparative Compound No. 2	8	15000	200	6000	2800
Comparative Example 6	Comparative Compound No. 3	8	15600	200	8000	4000

(Example 19)

A device with the structure shown in Fig. 3 was prepared.

The hole transporting layer 5 was formed on the transparent conductive supporting substrate in the same manner as in Example 1.

In addition, the Ir complex represented by the following structural formula and the carbazole compound represented by the following structural formula (polymerization ratio of 5:100) were deposited by a vacuum evaporation process in a thickness of 20 nm to form the light emitting layer 3. As for the conditions, the degree of the vacuum at the evaporation was  $1.0 \times 10^{-4}$  Pa and the film formation rate was 0.2-0.3 nm/sec.





Moreover, Exemplary Compound No. 5 was deposited in a thickness of 40 nm to form the electron transporting layer 6. As for the conditions, the degree of the vacuum at the evaporation was  $1.0 \times 10^{-4}$  Pa and the film formation rate was 0.2-0.3 nm/sec.

The device was then sealed after the cathode 4 was formed in the same manner as in Example 1.

When the thus obtained device was applied with a DC voltage of 10 V using the ITO electrode (anode 2) as a positive electrode and the Al-Li electrode (cathode 4) as a negative electrode, the current passed through the device at a current density of 20.0 mA/cm<sup>2</sup> and emission of green light was observed at a luminance of 6,800 cd/m<sup>2</sup>.

Furthermore, when the voltage was applied for 100 hours while maintaining the current density at 6.0 mA/cm<sup>2</sup>, the initial luminance of 1,300 cd/m<sup>2</sup> dropped to 1,150 cd/m<sup>2</sup> after 100 hours, exhibiting only a small reduction of luminance.

(Examples 20 to 31)

Devices were prepared and evaluated in the same manner as in Example 19 with the exception that Exemplary Compounds shown in Table 3 were used instead of Exemplary Compound No. 5. The results are shown in Table 3.

(Comparative Examples 7 to 9)

Devices were prepared and evaluated in the same

manner as in Example 19 with the exception that Comparative Compound Nos. 1 to 3 were used instead of Exemplary Compound No. 5. The results are shown in Table 3.

Table 3

Example No.	Exemplary Compound No.	Initial		Durability		
		Applied Voltage (v)	Luminance (cd/m <sup>2</sup> )	Current Density (mA/cm <sup>2</sup> )	Initial Luminance (cd/m <sup>2</sup> )	Luminance after 100 hours (cd/m <sup>2</sup> )
Example 19	5	10	6800	6.0	1300	1150
Example 20	4	10	5400	6.0	950	700
Example 21	6	10	6750	6.0	1298	1130
Example 22	7	10	6580	6.0	1050	880
Example 23	12	10	6510	6.0	1040	800
Example 24	13	10	6410	6.0	1056	800
Example 25	14	10	6680	6.0	1110	900
Example 26	18	10	5800	6.0	903	690
Example 27	19	10	5600	6.0	960	700
Example 28	20	10	6730	6.0	1220	980
Example 29	24	10	5800	6.0	960	700
Example 30	25	10	5980	6.0	970	610
Example 31	28	10	6680	6.0	990	710
Comparative Example 7	Comparative Compound No. 1	10	840	6.0	500	230
Comparative Example 8	Comparative Compound No. 2	10	500	6.0	300	150
Comparative Example 9	Comparative Compound No. 3	10	1300	6.0	800	300

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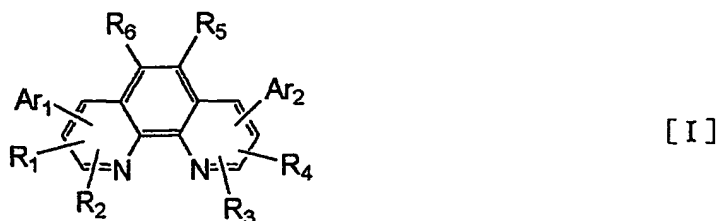
As described above by illustrating embodiments and examples, the organic light emitting devices using the phenanthroline compounds represented by the general formulas [I] to [III] provide the emission  
5 having a high luminance at a low applied voltage and are also excellent in durability. Particularly, the organic layers comprising the phenanthroline compounds of the present invention are excellent as an electron transporting layer as well as a light  
10 emitting layer.

Moreover, it is possible to prepare the devices by using a vacuum evaporation process, casting process or the like, and the devices having a large area can be prepared easily at a relatively low cost.

15

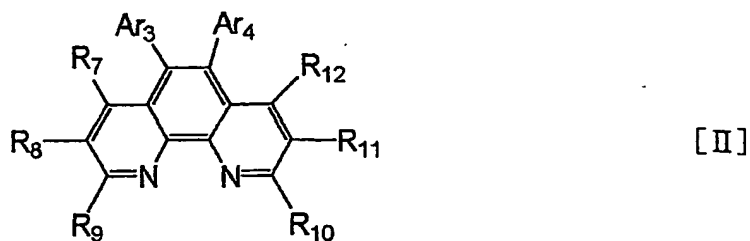
## CLAIMS

1. A phenanthroline compound represented by the general formula [I]:



5 (wherein R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, R<sub>4</sub>, R<sub>5</sub> and R<sub>6</sub> are the same or different and each is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an  
 10 unsubstituted or substituted heterocyclic group, and a halogen atom; and Ar<sub>1</sub> and Ar<sub>2</sub> are the same or different and each is selected from an unsubstituted or substituted fluorenyl group, an unsubstituted or substituted fluoranthenyl group, an unsubstituted or  
 15 substituted perylenyl group, and an unsubstituted or substituted carbazolyl group).

2. A phenanthroline compound represented by the general formula [II]:



20 (wherein R<sub>7</sub>, R<sub>8</sub>, R<sub>9</sub>, R<sub>10</sub>, R<sub>11</sub> and R<sub>12</sub> are the same or

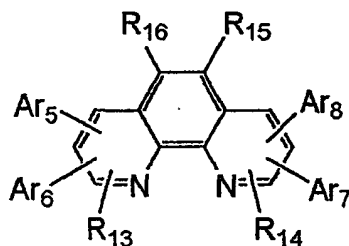
- 52 -

different and each is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an

5 unsubstituted or substituted heterocyclic group, and a halogen atom; and Ar<sub>3</sub> and Ar<sub>4</sub> are the same or different and each is selected from an unsubstituted or substituted fluorenyl group, an unsubstituted or substituted fluoranthenyl group, an unsubstituted or

10 substituted perylenyl group, and an unsubstituted or substituted carbazolyl group).

3. A phenanthroline compound represented by the general formula [III]:



[III]

15

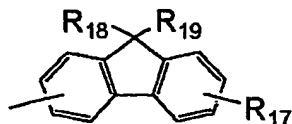
(wherein R<sub>13</sub>, R<sub>14</sub>, R<sub>15</sub> and R<sub>16</sub> are the same or different and each is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an

20 unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, and a halogen atom; and Ar<sub>5</sub>, Ar<sub>6</sub>, Ar<sub>7</sub> and Ar<sub>8</sub> are the same or different and each is selected from an

unsubstituted or substituted fluorenyl group, an unsubstituted or substituted fluoranthenyl group, an unsubstituted or substituted perylenyl group, and an unsubstituted or substituted carbazolyl group).

5

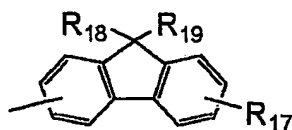
4. The phenanthroline compound according to claim 1, wherein the fluorenyl group is represented by the general formula [IV]:



[IV]

10 (wherein R<sub>17</sub> is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, a  
15 substituted amino group, a cyano group, and a halogen atom; and R<sub>18</sub> and R<sub>19</sub> are the same or different and each is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an  
20 unsubstituted or substituted aryl group, and an unsubstituted or substituted heterocyclic group).

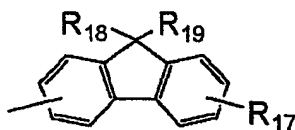
5. The phenanthroline compound according to claim 2, wherein the fluorenyl group is represented  
25 by the general formula [IV]:



[IV]

(wherein R<sub>17</sub> is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, a substituted amino group, a cyano group, and a halogen atom; and R<sub>18</sub> and R<sub>19</sub> are the same or different and each is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, and an unsubstituted or substituted heterocyclic group).

6. The phenanthroline compound according to claim 3, wherein the fluorenyl group is represented by the general formula [IV]:



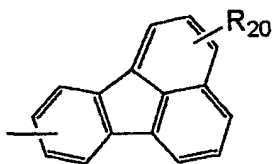
[IV]

(wherein R<sub>17</sub> is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, a



substituted amino group, a cyano group, and a halogen atom; and  $R_{18}$  and  $R_{19}$  are the same or different and each is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an  
5 unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, and an unsubstituted or substituted heterocyclic group).

7. The phenanthroline compound according to  
10 claim 1, wherein the fluoranthenyl group is represented by the general formula [V]:

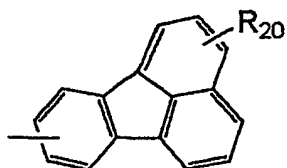


[V]

(wherein  $R_{20}$  is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an  
15 unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, a substituted amino group, a cyano group, and a halogen atom).

20

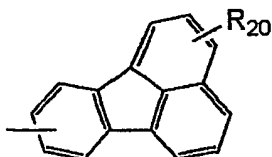
8. The phenanthroline compound according to claim 2, wherein the fluoranthenyl group is represented by the general formula [V]:



[V]

(wherein R<sub>20</sub> is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an  
 5 unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, a substituted amino group, a cyano group, and a halogen atom).

10            9.    The phenanthroline compound according to claim 3, wherein the fluoranthenyl group is represented by the general formula [V]:

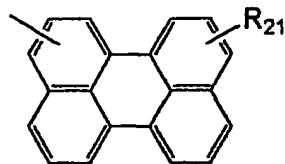


[V]

(wherein R<sub>20</sub> is selected from a hydrogen atom, an  
 15 unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, a substituted amino group, a cyano group, and a halogen  
 20 atom).

10.    The phenanthroline compound according to claim 1, wherein the perylenyl group is represented

by the general formula [VI]:

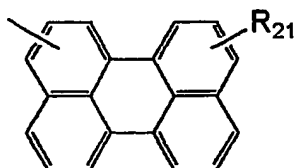


[VI]

(wherein R<sub>21</sub> is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, a substituted amino group, a cyano group, and a halogen atom).

10

11. The phenanthroline compound according to claim 2, wherein the perylenyl group is represented by the general formula [VI]:



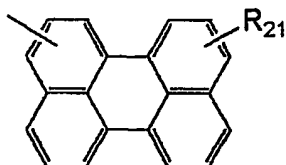
[VI]

15 (wherein R<sub>21</sub> is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, a substituted amino group, a cyano group, and a halogen atom).

20

12. The phenanthroline compound according to

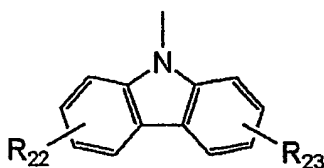
claim 3, wherein the perylenyl group is represented by the general formula [VI]:



[VI]

(wherein R<sub>21</sub> is selected from a hydrogen atom, an  
5 unsubstituted or substituted alkyl group, an  
unsubstituted or substituted aralkyl group, an  
unsubstituted or substituted aryl group, an  
unsubstituted or substituted heterocyclic group, a  
substituted amino group, a cyano group, and a halogen  
10 atom).

13. The phenanthroline compound according to claim 1, wherein the carbazolyl group is represented by the general formula [VII]:



[VII]

15 (wherein R<sub>22</sub> and R<sub>23</sub> are the same or different and  
each is selected from a hydrogen atom, an  
unsubstituted or substituted alkyl group, an  
unsubstituted or substituted aralkyl group, an  
20 unsubstituted or substituted aryl group, an  
unsubstituted or substituted heterocyclic group, a  
substituted amino group, a cyano group, and a halogen  
atom).

14. The phenanthroline compound according to claim 2, wherein the carbazolyl group is represented by the general formula [VII]:



5 (wherein R<sub>22</sub> and R<sub>23</sub> are the same or different and each is selected from a hydrogen atom, an unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an  
10 unsubstituted or substituted heterocyclic group, a substituted amino group, a cyano group, and a halogen atom).

15 15. The phenanthroline compound according to claim 3, wherein the carbazolyl group is represented by the general formula [VII]:



(wherein R<sub>22</sub> and R<sub>23</sub> are the same or different and each is selected from a hydrogen atom, an  
20 unsubstituted or substituted alkyl group, an unsubstituted or substituted aralkyl group, an unsubstituted or substituted aryl group, an unsubstituted or substituted heterocyclic group, a

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substituted amino group, a cyano group, and a halogen atom).

16. An organic light emitting device  
5 comprising a pair of electrodes consisting of an anode and a cathode, and a layer comprising an organic compound comprising the phenanthroline compound set forth in claim 1, interposed between the pair of electrodes.

10

17. An organic light emitting device  
comprising a pair of electrodes consisting of an anode and a cathode, and a layer comprising an organic compound comprising the phenanthroline  
15 compound set forth in claim 2, interposed between the pair of electrodes.

18. An organic light emitting device  
comprising a pair of electrodes consisting of an  
20 anode and a cathode, and a layer comprising an organic compound comprising the phenanthroline compound set forth in claim 3, interposed between the pair of electrodes.

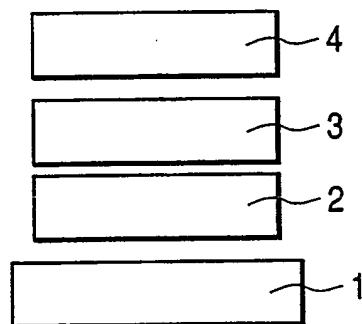
25 19. The organic light emitting device  
according to claim 18, wherein the layer comprising the organic compound comprising the phenanthroline

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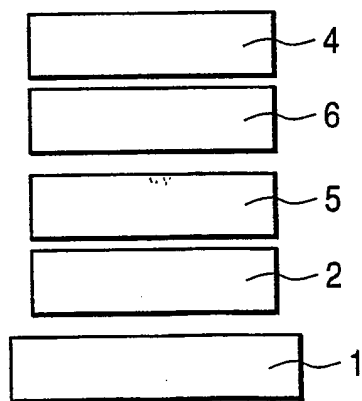
compound functions as an electron transporting layer or a light emitting layer.

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**FIG. 1**



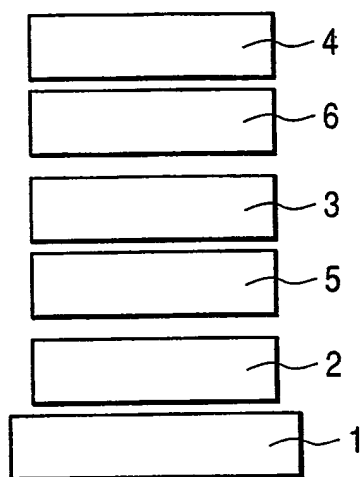
**FIG. 2**



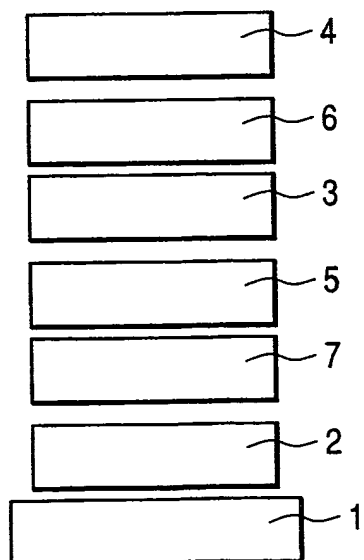


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**FIG. 3**

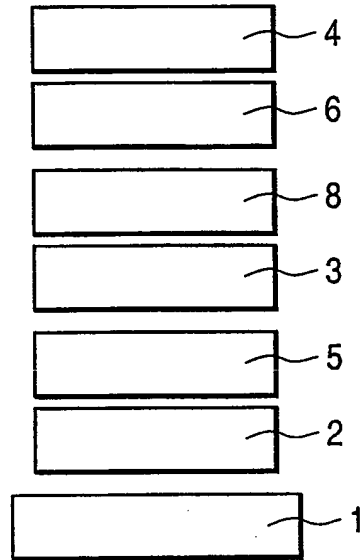


**FIG. 4**

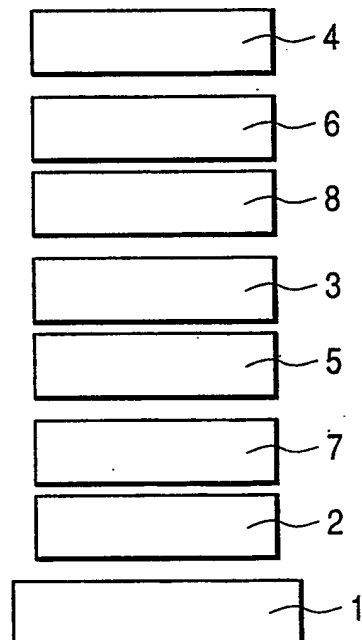


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**FIG. 5**



**FIG. 6**



## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP03/11485

## A. CLASSIFICATION OF SUBJECT MATTER

Int.Cl<sup>7</sup> C07D471/04, C09K11/06, H05B33/14, H05B33/22

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Int.Cl<sup>7</sup> C07D471/04, C09K11/06, H05B33/14, H05B33/22

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched  
 Japanese Utility Model Gazette 1926-1996, Japanese Publication of Unexamined Utility Model Applications 1971-2001, Japanese Registered Utility Model Gazette 1994-2001, Japanese Gazette Containing the Utility Model 1996-2001

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

CAOLD (STN), CAPLUS (STN), REGISTRY (STN)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X Y	EP 1097980 A2 (SONY CORPORATION) 2001.05.09, page 30-31 & EP 1097980 A3 & JP 2001-131174 A	1,16,19 2-15,17,18
X Y	EP 1097981 A2 (SONY CORPORATION) 2001.05.09, page 4, page 29-30 & EP 1097981 A3 & JP 2001-135482 A & US 6524728 A	1,2,16,17, 19 3-15,18
Y	JP 2001-267080 A (TORAY INDUSTRIES, INC.) 2001.09.28, page 5-6, page 8 (Family: none)	1-19
Y	ITO, T., "Synthesis of phenanthroline derivatives and their application to organic EL devices" Polymer Preprints, Japan, (2002.09.18), VOL.51, NO.11, pp.2859-2860	1-19
Y	JP 2002-050481 A (TORAY INDUSTRIES, INC.) 2002.02.15, page 4 (Family: none)	1-19

☐ Further documents are listed in the continuation of Box C.☐ See patent family annex.

## \* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&amp;" document member of the same patent family

Date of the actual completion of the international search

07.11.03

Date of mailing of the international search report

25.11.03

Name and mailing address of the ISA/JP

Japan Patent Office

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Authorized officer

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Telephone No. +81-3-3581-1101 Ext. 3492



4P

9837

# INTERNATIONAL SEARCH REPORT

International application No.  
**PCT/JP 03/11485**

## Box I Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
  
2. ☐ Claims Nos.:  
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
  
3. ☐ Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box II Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

**See the attached paper.**

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☒ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
  
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.  
☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT  
Information on patent family members

International application No.

PCT/JP03/11485

The invention is composed of the following 4 groups;  
phenanthroline compounds substituted fliorennyl group (claims 1-6,16-19),  
phenanthroline compounds substituted flioranthenyl group (claims 1-3,7-9,16-19),  
phenanthroline compounds substituted perylenyl group (claims 1-3,10-12,16-19),  
and phenanthroline compounds substituted carbazolyl group (claims 1-3,13-19 ).

Claims 1-19 have the phenanthroline moiety useful for an organic light emitting device in common, however, it does not appear to be novel, as it is disclosed in D1-4 (D1: EP 1097980 A2, D2: EP 1097981 A2, D3: JP 2001-267080 A, D4: Polymer Preprints, Japan, (2002.09.18), VOL.51, NO.11, pp.2859-2860). In other words, the common moiety above-mentioned is within the reach of prior art, and it is not considered as "special technical features".

Consequently, there is no technical relationship among those inventions involving one or more of the same or corresponding technical features. Therefore, these groups of invention are not linked so as to form a single general inventive concept.

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